# Progress in the Development of the MISTE Flight Experiment

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The MISTE flight experiment plans to measure the specific heat at constant volume,  $C_V$ , and isothermal susceptibility,  $\chi_T$ , near the  $^3$ He liquid-gas critical point. Precision ground-based experiments have been performed in the crossover region away from the critical point in preparation for this flight. A new method for precisely determining the critical temperature is being evaluated and will be presented. A sweep electrostriction method was also demonstrated for obtaining the isothermal susceptibility close to the critical point. We have been able to demonstrate that the chemical potential can be obtained from these electrostriction measurements. Pressure versus density measurements along isotherms below the critical temperature were performed to determine the isothermal susceptibility along the coexistence curve. These measurements are compared to susceptibility data obtained along the critical isochore above the transition.

#### 1. INTRODUCTION

The MISTE flight experiment plans to perform heat capacity at constant volume,  $C_V$ , isothermal susceptibility,  $\chi_T$ , and PVT measurements in the same experimental cell near the liquid-gas critical point of  $^3$ He. These experiments, performed in a microgravity environment, will provide measurements in the asymptotic region two decades in reduced temperature closer to the transition than obtained on earth. The fluctuation-induced constant-volume heat capacity along the critical isochore and the isothermal susceptibility along the critical isochore and coexistence curve are expected to satisfy the following theoretical expressions:

$$C_V^{\pm *} = (T_c \rho_c / P_c) C_V^{\pm}$$
  
=  $A_0^{\pm} |t|^{-\alpha} [1 + A_1^{\pm} |t|^{\Delta_s} + ...] + B_{cr},$  (1)

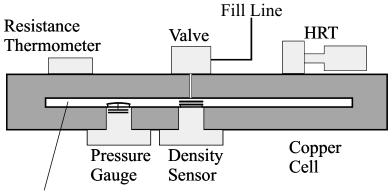
$$\chi_T^{\pm *} = (P_c/\rho_c^2)\chi_T^{\pm} 
= \Gamma_0^{\pm}|t|^{-\gamma}[1 + \Gamma_1^{\pm}|t|^{\Delta_s} + ...],$$
(2)

where  $\alpha \simeq 0.11$  and  $\gamma \simeq 1.24$  are universal critical exponents and  $A_0^{\pm}$  and  $\Gamma_0^{\pm}$  are system-dependent critical amplitudes. The superscripts "+" and "–" correspond to positive and negative reduced temperatures  $t \equiv (T - T_c)/T_c$ , respectively. The isothermal susceptibility along the coexistence curve is given by  $\chi_T^{-*}$ . The system-dependent critical parameters are  $T_c$ ,  $\rho_c$ , and  $P_c$ , and  $P_c$  is a fluctuation-induced, constant term. The confluent singularity expansion in the brackets includes an independent universal correction-to-scaling exponent,  $\Delta_s = 0.52\pm0.02$  and system-dependent amplitudes  $\Delta_1^{\pm}$  and  $\Delta_2^{\pm}$  and  $\Delta_3^{\pm}$  and  $\Delta_3^{\pm}$  and  $\Delta_3^{\pm}$  and  $\Delta_3^{\pm}$  and  $\Delta_3^{\pm}$  and  $\Delta_3^{\pm}$  are universal critical exponents and  $\Delta_3^{\pm}$  are universal critical exponents and  $\Delta_3^{\pm}$  and  $\Delta_3^{\pm}$  are universal critical exponents and  $\Delta_3^{\pm}$  are universal exponents.

Critical phenomena theories can predict critical exponents and universal amplitude ratios. However, an exact determination of the asymptotic region cannot be made theoretically since the leading critical amplitudes and the amplitudes associated with correction-to-scaling confluent singularities are system dependent. The MISTE flight experiment should permit an accurate determination of the leading system-dependent asymptotic critical amplitudes for <sup>3</sup>He. A knowledge of these asymptotic amplitudes will permit a more accurate analysis of crossover measurements farther away from the transition. Ground-based studies are now being performed in the crossover region<sup>2</sup> in preparation for this future flight experiment. The results of some of these studies will be reported in this paper.

#### 2. GROUND-BASED EXPERIMENTS

Ground-based measurements were performed in a flat pancake fluid cell shown in Fig. 1. The cell temperature was measured using a GdCl<sub>3</sub> high resolution thermometer (HRT) with a sensitivity of  $\sim 1$  nK near the <sup>3</sup>He critical point ( $T_{\rm c}=3.31$  K). The density sensor was a parallel plate capacitor situated half way between the top and bottom of the cell. The density was determined from the measurement of the dielectric constant using the Clausius-Mossotti equation. A Straty-Adams type pressure sensor was also situated at the midplane of the cell. This sensor consisted of a parallel plate



Sample Height = 0.05 cm, Diameter = 11.2 cm

Fig. 1. Schematic of ground-based cell for measuring heat capacity and susceptibility.

capacitor with one plate attached to a flexible diaphragm that sensed pressure changes in the cell. This experimental configuration has the advantage that pressure, density, and temperature data can be simultaneously obtained in the same cell while heat capacity and susceptibility measurements were being performed.

The isothermal susceptibility,  $\chi_T = \rho(\partial \rho/\partial P)_T$ , was measured along isotherms both above and below the critical temperature. This was achieved by initially overfilling the cell and then slowly removing fluid from the cell and measuring the density and pressure as a function of time. Susceptibility data were obtained from the slope of P versus  $\rho$  curves in the reduced temperature range of  $6 \times 10^{-5} < |t| < 10^{-1}$ . The density of the susceptibility maximum approaches the critical density,  $\rho_c$ , as  $T \to T_c$ . After completing the susceptibility measurements, the low temperature valve was closed at the critical density. Heat-capacity measurements were then performed using a pulse technique in the single and two-phase regions over the range  $5 \times 10^{-4} < |t| < 10^{-1}$ . Drift heat-capacity measurements were also performed close to the transition. These new  $C_V$  and  $\chi_T$  data agreed with earlier measurements from Horst Meyer's group<sup>3-6</sup> over the same temperature range of overlap.

# 3. Determination of the critical temperature

In order to successfully analyze experimental measurements near a critical point, it is important to determine the critical temperature as precisely

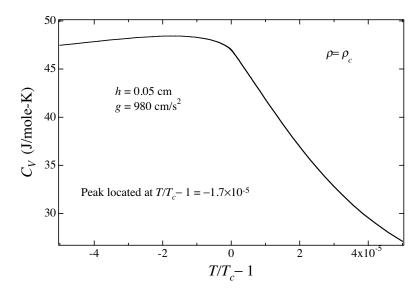


Fig. 2. Restricted cubic model prediction for the <sup>3</sup>He heat capacity in a 0.05 cm high cell along the critical isochore in 1g. A change in slope is predicted at the transition temperature.

as possible. There are several methods for finding  $T_c$ . One approach is to have  $T_c$  be a fitting parameter when analyzing data. However, the accuracy of the resultant  $T_c$  will depend on the model chosen and closeness of the data to the transition. We have used this approach with susceptibility measurements above the transition<sup>7</sup> to obtain  $T_c = 3.315533 \,\mathrm{K}$ . This  $T_c$  will be used in analyzing the susceptibility measurements presented later in this paper. One can also attempt to use some characteristic signature of the transition. For example, it is known that the time constant for equilibrium in performing heat-capacity measurements is much greater in the two-phase region. Unfortunately, in ground-based measurements, the gravity induced density gradient smears out this sudden change near the transition.

We have recently been investigating a new slow-drift heat-capacity approach for determining the transition temperature. For experimental cells of small vertical height in a gravitational field, theory predicts an experimentally measurable change in the temperature derivative of  $C_V$  at  $T_c$  with a heat capacity maximum occurring in the two-phase region. Figure 2 shows the restricted cubic model<sup>8</sup> prediction for the <sup>3</sup>He heat capacity in our 0.05 cm high cell along the critical isochore in 1g. We see a change in slope occurring at the transition and a peak value located at a reduced temperature of  $t \approx -1.7 \times 10^{-5}$  below  $T_c$ . Figure 3 shows an example of our experimental

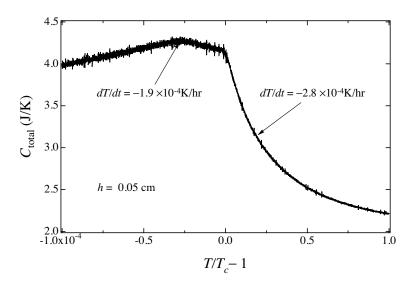


Fig. 3. Experimental determination of the transition temperature using the <sup>3</sup>He heat capacity anomally along the critical isochore in 1g.

measurements performed during a slow drift down through the transition. There is a kink in the data that we associate with the transition temperature  $T_c$ . The transition temperature can be determined to better than  $\pm 15 \mu \rm K$ . The peak in these data is at  $t \approx -2.3 \times 10^{-5}$ , which is close to the theoretically predicted value. Additional measurements are planned at even slower drift rates to make sure we have this level of reproducibility in determining  $T_c$ .

# 4. Chemical potential

An electrostriction technique was previously developed to perform susceptibility measurements within  $t<10^{-4}$ . This technique is based on the fact that an electric field gradient can produce a pressure gradient within a dielectric fluid  $(\delta P \propto E^2)$  that in turn induces a density change. Our approach is to apply a dc voltage across a parallel plate capacitor to produce a uniform electric field within the density sensor capacitor gap. The density change  $\delta \rho$  is obtained at several voltages and  $\delta \rho/\delta P$  is determined in the limit of zero voltage. We have been evaluating drift electrostriction measurements using a fixed dc voltage across the gap. Figure 4 shows measurements of the density change between the applied dc voltage and zero dc voltage. The drift rates were  $\approx -4 \times 10^{-4}$  K/hr. The susceptibility at any

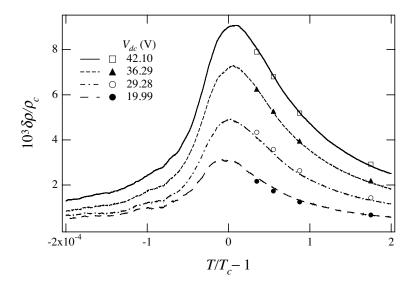


Fig. 4. Density change versus temperature for electrostriction drift (lines) and equilibrium (symbols) measurements at various constant applied dc bias voltages.

given temperature can be determined by extrapolating the density values obtained from these curves to zero dc voltage. We also performed equilibrium measurements, shown by symbols, at a few reduced temperatures to see if this drift rate was slow enough. Good agreement was obtained between the drift and equilibrium measurements down to about a reduced temperature of  $t = 5 \times 10^{-5}$ . The MISTE flight experiment plans to reach a reduced temperature of  $10^{-6}$  that will require even slower drift rates.

We have realized that this electrostriction technique can be used to conveniently determine the chemical potential difference that can be written as  $d\mu = -sdT + dP/\rho$ . Under isothermal conditions the integration of this expression reduces to

$$\mu(\rho_2, T) - \mu(\rho_1, T) = \int_{P(\rho_1, T)}^{P(\rho_2, T)} dP/\rho, \qquad (3)$$

where  $\rho_1$  is the ambient density outside the capacitor gap and  $\rho_2$  is the density inside the gap. Since the pressure gradient induced by an electric field gradient is given by

$$\nabla P = (\epsilon_0 \rho / 2) \nabla (E^2(\partial \epsilon / \partial \rho)), \tag{4}$$

we can write the chemical potential difference between two densities, Eq. (3),

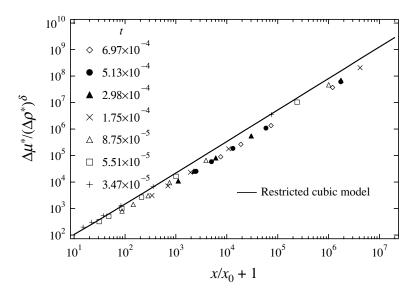


Fig. 5. Comparison of electrostriction measurements to theoretically predicted universal curve for the chemical potential.

as

$$\mu(\rho_2, T) - \mu(\rho_1, T) = (3\epsilon_0 \zeta_c \rho_c / 2(\rho_c - \zeta_c \rho_2)^2) E^2, \tag{5}$$

where  $\zeta_c = 4\pi\alpha'/3$  with  $\alpha'$  being the polarizability of the fluid.

In the asymptotic region, scaling theory predicts the chemical potential difference from its critical value can be scaled by a universal function h

$$(\mu^*(\rho, T) - \mu^*(\rho_c, T))/(\Delta \rho |\Delta \rho|^{\delta - 1}) = Dh(x/x_0),$$
(6)

where  $\mu^* = (\rho_c/P_c)\mu$ ,  $x = t/(|\Delta\rho|)^{1/\beta}$ , and  $x_0 = 1/B_0^{1/\beta}$ . The reduced density  $\Delta\rho \equiv \rho/\rho_c - 1$ , and  $\beta \simeq 0.326$  is the critical exponent and  $B_0$  the critical amplitude that define the shape of the coexistence curve. D is the critical amplitude associated with the divergence of the pressure with density along the critical isotherm. By setting the ambient density  $\rho_1$  equal to the critical density in Eq. (5), we obtain the following expression for the universal function h

$$Dh(x/x_0) = (3\epsilon_0 \zeta_c \rho_c / 2(\rho_c - \zeta_c \rho_2)^2)(E^2 / (\Delta \rho)^{\delta}). \tag{7}$$

The right side of this equation can be determined experimentally since we measure the density  $\rho_2$  and calculate  $\Delta \rho$  and the electric field. Figure 5 is a plot of the universal curve. The solid line is the prediction from the restricted

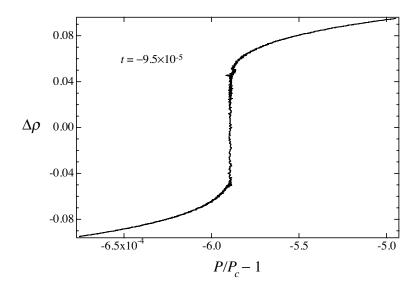


Fig. 6. Pressure versus density along an isotherm below  $T_c$ .

cubic model<sup>8</sup> and the data points come from equilibrium electrostriction measurements at various temperatures. We do not expect to have a good fit between theory and experiment since most of the data points are not in the asymptotic region. However, we anticipate a much more stringent test of this scaling theory prediction coming from the MISTE flight experiment where measurements will be performed much closer to the transition.

# 5. Susceptibility measurements

The isothermal susceptibility along the coexistence curve can also be obtained from pressure versus density measurements as stated above. Figure 6 shows an example of P versus  $\rho$  measurements at a reduced temperature of  $t = -9.5 \times 10^{-5}$ . The break in  $(\partial \rho/\partial P)_T$  at a reduced density  $\Delta \rho \approx 5\%$  indicates the onset of the two-phase region in which the pressure remains constant until the gas side of the coexistence curve is reached at  $\Delta \rho \approx -5\%$ . The susceptibility at the coexistence curve is obtained from the slope of this curve as it enters and exits the two-phase region. The coexistence curve densities at this temperature can also be determined to within  $\pm$  0.1% from such breaks in the slope. This method for determining the susceptibility and coexistence curve will become less precise as the transition is approached since the change in slope will be less pronounced.

The experimentally determined susceptibility along the critical isochore

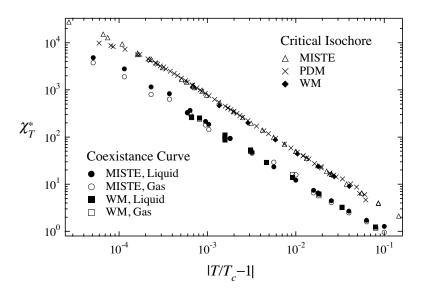


Fig. 7. Susceptibility measurements along the critical isochore above  $T_c$  and along the coexistence curve below  $T_c$ . Also included are the data from Wallace and Meyer (WM), and Pittman, Doiron and Meyer (PDM).

and coexistence curve are shown in Fig. 7. Earlier measurements from Wallace and Meyer<sup>3</sup>, and Pittman, Doiron, and Meyer<sup>4</sup> have also been included. The MISTE measurements along the coexistence curve extend over a decade closer to the transition than the earlier data. We see good agreement between the various measurements in the regions of overlap. The susceptibility along the coexistence curve and critical isochore seem to have the same slope in this log-log plot, with the coexistence curve susceptibility having a smaller magnitude. To estimate the difference in magnitude between the susceptibility data above and below the transition, the critical isochore data were divided by 3.7 to approximately overlap the coexistence curve data. This is the same value used by Wallace and Meyer in their earlier work.<sup>3</sup> These normalized data are shown in Fig. 8. It is surprising to see that all the data sets are consistent with each other over three decades in reduced temperature. This factor of 3.7 can be considered as an effective ratio of the critical amplitudes  $(\Gamma_0^+/\Gamma_0^-)_{\text{eff}}$  in the crossover region. Critical phenomena theories 10 predict a ratio  $\Gamma_0^+/\Gamma_0^- = 4.95$  in the asymptotic region. Since most of these ground-based susceptibility data are outside the asymptotic region, additional measurements from the MISTE flight experiment are needed closer to the transition to test this theoretical asymptotic prediction and to obtain a better understanding of the observed crossover behavior.

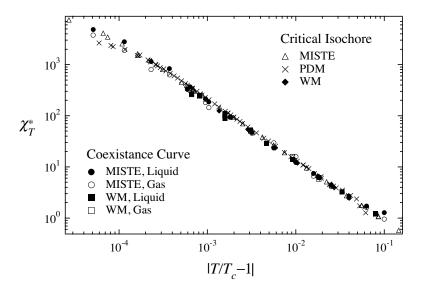


Fig. 8. Normalized susceptibility measurements. Critical isochore data above  $T_c$  in Fig. 7 were divided by 3.7 to overlay on coexistence data below  $T_c$ .

# 6. FUTURE STUDIES

The recent heat capacity and susceptibility measurements need to be analyzed using accurate theoretical crossover models in order to obtain a better understanding of thermodynamic behavior farther away from the transition. We began this process by analyzing our susceptibility data along the critical isochore above the transition using a field-theoretical Renormalization-Group  $\phi^4$  approach<sup>11</sup> recently adapted to the O(1) universality class.<sup>12</sup> The results of this initial study<sup>7</sup> suggested that the  $\phi^4$  model would be a useful tool to investigate thermodynamic quantities measured along the critical isochore and coexistence curve. More recently, we have used this model to successfully analyze our heat-capacity data along the critical isochore.<sup>13</sup> The MISTE susceptibility and heat-capacity data have also been analyzed successfully using a new crossover parametric equation-of-state.<sup>13</sup> Further analyses of thermodynamic measurements throughout the critical region are planned using both the crossover parametric equation-of-state and  $\phi^4$  models.

#### ACKNOWLEDGMENTS

The research described in this article was carried out at the Jet Propulsion Laboratory, California Institute of Technology, under contract with the National Aeronautics and Space Administration. The authors are indebted to Dr. M. Weilert for supporting the experimental measurements and data analysis.

#### REFERENCES

- 1. R. Guida and J. Zinn-Justin, J. Phys. A: Math. Gen. 31, 8103 (1998).
- 2. M. Barmatz, MISTE Science Requirements Document, Tech. Rep. JPL D-17083, JPL (1999).
- 3. B. Wallace and H. Meyer, Phys. Rev. A 5, 953 (1972).
- 4. C. Pittman, T. Doiron, and H. Meyer, Phys. Rev. B 20, 3678 (1979).
- C. C. Agosta, S. Wang, L. H. Cohen, and H. Meyer, J. Low Temp. Phys. 67, 237 (1987).
- 6. R. G. Brown and H. Meyer, Phys. Rev. A 6, 364 (1972).
- 7. I. Hahn, F. Zhong, M. Barmatz, R. Haussmann, and J. Rudnick (to be published).
- 8. F. Zhong and H. Meyer, Phys. Rev. E 51, 3223 (1995).
- 9. M. Barmatz, F. Zhong, and I. Hahn, Physica B 284-288, 206 (2000).
- 10. M. E. Fisher and S.-Y. Zinn, J. Phys. A: Math. Gen. 31, L629 (1998).
- 11. R. Schloms and V. Dohm, Nuclear Physics **B328**, 639 (1989).
- 12. R. Haussmann (private communication).
- 13. M. Barmatz, I. Hahn, F. Zhong, M. A. Anisimov, and V. A. Vaktang (to be published).